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EVALUATION OF FILTERS USING SPECTROFLUORIMETRY AND CNC METHODS

H. Leung - Engineer
M. MacKay - Student
Environmental Science Section
Chemical Research Department

ABSTRACT

Test uranine aerosols are used to study aerosol transport in the stacks of the nuclear generating stations. The present method of analysis involves the collection of aerosol on filters followed by analysis for uranine using a spectrofluorimetric technique. This procedure is labour-intensive and involves significant time delays. An alternative approach involving the use of real-time aerosol detection using a Condensation Nucleus Counter (CNC) is under consideration. In this study, the two methods were compared in the laboratory. It was determined that the two methods give essentially identical results for the collection efficiency of a glass fibre test filter. It is, therefore, recommended that the CNC method be used instead of the spectrofluorimetric method for aerosol transport studies at the nuclear stations.

1.0 INTRODUCTION

At the request of Radioactive Management and Environmental Protection, we have determined the efficiency of glass fibre filters using two independent methods - one utilizing spectrofluorimetry and the other, a condensation nucleus counter (CNC). Spectrofluorimetry methods are presently used for particle transport studies in the stacks of Ontario Hydro's nuclear generating stations.

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Known quantities of uranine particles (sodium fluorescein) are injected into the stack and their concentrations are obtained by spectrofluorimetric measurement of uranine collected on filters. The spectrofluorimetric method is sensitive at very low uranine concentrations (ppt range), however this method requires laboratory analysis of collected samples, and is quite time-consuming. On the other hand, the CNC method provides in-situ and continuous measurements of particle concentration. This method is not limited to uranine but can be used for any liquid or solid particles as long as they fall in the submicrometer size range. The CNC method requires a continuous flow of particles through the counter, at near ambient conditions. The number concentrations are measured by an optical counter.

The CNC method has obvious advantages over the spectrofluorimetry method. The possibility of substituting the CNC method for in-stack particle measurements was therefore investigated in this study. We compared the filter efficiency results of these two methods using the Whatman GF/B glass fibre filter. The spectrofluorimetric method yields filter collection efficiency based on mass measurement, while the CNC method provides efficiency based on count measurement. These two measurements may differ slightly, depending on the particle size distribution, but for monodispersed aerosols this difference is negligible.

2.0 EXPERIMENTAL SETUP

The experimental setup is shown in Figure 1. It consists of a six-jet atomizer, diffusion dryer, neutralizer (if needed), filter holder assembly (Gelman stainless steel in-line model 2220, effective filtration area 9.62 cm^2), electrostatic classifier (EC) and condensation nucleus counter (CNC). The EC is not required for field study, but was used in this work since the EC/CNC combination gives additional information on particle size distribution and filter volume efficiency. The filter volume efficiency was obtained indirectly from the particle count distribution. A spectrofluorimeter was used for uranine analysis. A brief description of the equipment is given below.

2.1 Six-jet Atomizer

A TSI model 9306 six-jet atomizer, on loan from the Analytical Services Section, was used to generate uranine particles. This instrument works on the principle of atomization of a solution of known uranine concentration into fine spray droplets by an air jet. Larger spray droplets are removed by impaction within the device. The volatile solvent evaporates, leaving the non-volatile dissolved substance behind as solid particles. This atomizer provides up to six jets. It has a built-in pressure regulator and gauge for control of input air pressure. It also

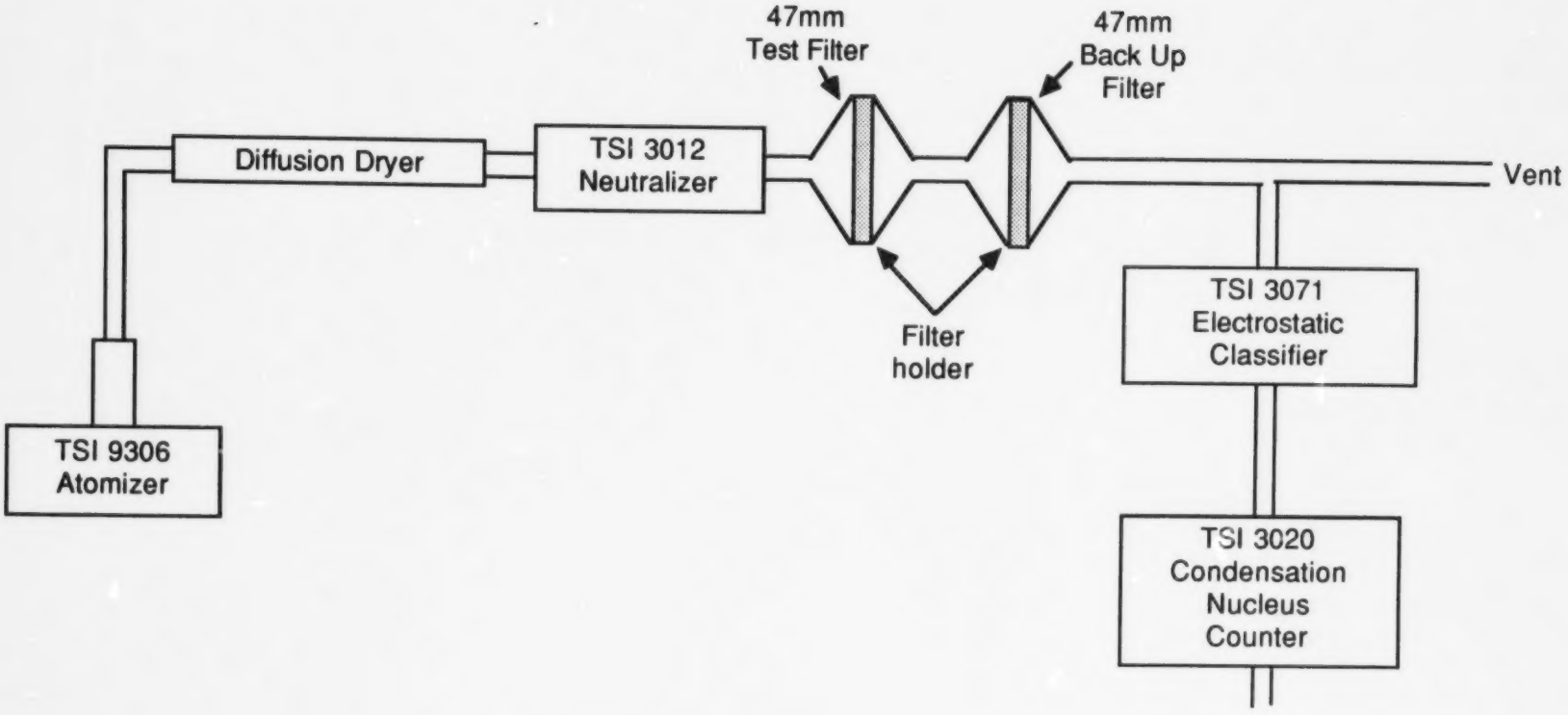

FIGURE 1
SCHEMATIC OF APPARATUS FOR FILTER TESTING

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contains a built-in rotameter and valve to regulate the flow of dilution air. The particle size, which depends on the solute concentration, ranges from about 0.04 to 4 μm. The particle generation rate depends on the input air pressure and the number of jets used. The maximum generation rate is about 7×10^{11} particles/min.

2.2 Diffusion Dryer

A diffusion dryer, similar to the TSI model 3062 diffusion dryer, was built. It consists of two concentric cylinders. The outer cylinder was made from Lucite material (5.72 cm ID) and the inner cylinder from 6-20 mesh wire screen (1.27 cm ID). The annular space between the inner and outer cylinders was filled with silica gel. Aerosol was dried as it passed through the inner cylinder and contacted the silica gel. The length of the dryer is 120 cm.

2.3 Neutralizer

A TSI model 3012 aerosol neutralizer was used to neutralize any remaining static charges on the particles. It contains a sealed krypton-85 source (2 mCi).

2.4 Electrostatic Classifier/Condensation Nucleus Counter

A TSI model 3071 electrostatic classifier was used to size particles, according to their mobility, over 39 size channels. The classifier can measure particle size in the range from 0.01 to 1.0 μm. The TSI model 3020 condensation nucleus counter was used to detect the particles as they exited from the classifier. The CNC can measure particle concentrations between 0.01 and 10^7 particles/cm^3.

2.5 Glass Fibre Filter

Whatman GF/B glass fibre filters (47-mm diameter) were studied. The characteristics of this filter are given in Table I/1/.

3.0 PROCEDURE

Referring to Figure 1, we see that the uranine aerosol was generated by the six-jet atomizer. The filter efficiency was then determined by two independent methods, spectrofluorimetry and CNC. The effects of flowrate and particle neutralization on filter efficiency were examined. The experiments were run according to the test matrix given in Table II. Each experimental condition was repeated once.

TABLE I

CHARACTERISTICS OF WHATMAN GF/B, GLASS FIBRE FILTER

ΔP^* (cm Hg)	1.0	3.0	5.0	10.0
V^+ (cm/s)	5.49	16.5	26.6	49.9
D_p^ϕ (μm)	Collection Efficiency (%)			
0.035	>99.99	>99.99	>99.99	>99.99
0.10	>99.99	>99.99	>99.99	>99.99
0.30	>99.99	>99.99	>99.99	>99.99
1.0	>99.99	>99.99	>99.99	>99.99

*Pressure drop
+Filter face velocity
$^\phi$Particle diameter

TABLE II

TEST MATRIX

	Flowrate		Neutralizer	
Run	25 L/min*	5 L/min+	With	Without
1-2	X		X	
3-4		X	X	
5-6	X			X
7-8		X		X

* Dilution flowrate + Aerosol flowrate which corresponds to a standard flowrate of 41.1 L/min and a filter face velocity of 71.2 cm/s

+ Aerosol only which corresponds to a standard flowrate of 8.1 L/min and a filter face velocity of 14.0 cm/s.

3.1 Aerosol Generation

Uranine aerosols were generated from a 10 g/L aqueous uranine solution using the TSI model 9306 six-jet atomizer. One jet was used and the air inlet pressure was kept at 25 psig. The total air flowrate was regulated using an external rotameter. The count mean diameter (CMD) and volume mean diameter (VMD) were about 0.11 μm and 0.35 μm, respectively, for uranine aerosols. The dilution air flowrate was set by changing the dilution air flow (built-in rotameter) and/or input total air flow (external rotameter). The particles were then dried by mixing the aerosol flow with the dilution air and by passing through the diffusion dryer. Any existing charges on the aerosol were removed by passing through the neutralizer.

3.2 Spectrofluorimetry Method

The aerosols were passed through the diffusion dryer-neutralizer (when used) - test filter - backup filter - EC-CNC train for about 9 minutes (Figure 1). The amounts of uranine on the test filter and backup filter were determined separately using the spectrofluorimeter. The EC/CNC setup was used to check if there were any particles breaking through the backup filter. We found there were very few particles coming through (close to background counts). The filter efficiency was determined by:

$$\text{Mass efficiency} = \frac{\text{weight of uranine on test filter}}{\text{total weight of uranine on test filter + backup filter}} \times 100\% \quad (1)$$

3.3 CNC Method

The aerosols were passed through the diffusion dryer-neutralizer (when used) - test filter - empty backup filter holder - EC-CNC train (Figure 1). The EC/CNC was adjusted to measure particle sizes from about 0.017 to 0.886 μm. The operating conditions were the same as in the spectrofluorimetric method. The total aerosol concentration after the test filter was obtained by the EC/CNC. The total aerosol concentration before the filter was measured in a similar manner but without the test filter. The filter efficiency was given by:

$$\text{Count efficiency} = \left(1 - \frac{\text{uranine concentration after test filter}}{\text{uranine concentration before test filter}}\right) \times 100\% \quad (2)$$

3.4 Spectrofluorimeter Technique

We followed the same procedure for analysis of uranine on the filters as outlined in the report by Curtis/2/. Uranine collected on the test filter and backup filter was each dissolved in 100 mL of buffer solution (0.1 M ammonium chloride/ammonium hydroxide, pH about 10.4). A known amount of the solution was further diluted with the buffer solution to reduce colour intensity to a level appropriate for the spectrofluorimeter (ppb range). We borrowed the Aminco-Bowman spectrophotofluorimeter from the Analytical Services Section to do the analysis. The excitation wavelength was set at 490 nm. The emitted fluorescence of each solution was scanned between 450 nm and 550 nm. The relative intensity reading at the peak (about 520 nm) was recorded and compared to a standard curve that was generated the same day. To generate the standard curve, four standards were prepared; 12.5, 25, 50, and 100 ppb. To ensure that degradation of the uranine by ultraviolet light was kept to a minimum, the samples were analyzed within two hours of preparation.

4.0 RESULTS

Typical uranine aerosol size spectra measured before and after passing through the Whatman GF/B filter are given in Figure 2. The CMD was about 0.11 $\pm$ 0.02 μm and the VMD was about 0.35 $\pm$ 0.04 μm (Table IV). (The spectrum after passing through the filter shows a spike near 0.8 μm particle size (Figure 2). This spike may be due to generation of a small number of particles at high voltage on the centre rod of the EC). A typical standard curve for the spectrofluorimetric analysis of uranine is given in Figure 3. Filter collection efficiencies measured with the spectrofluorimetric and CNC methods are given in Tables III and IV, respectively. Table V compares the filter efficiency data derived by the two methods.

5.0 DISCUSSION

We compared the filter collection efficiency using two independent methods - spectrofluorimetry and CNC. The results from these two methods agree very well, within 0.1% (Table V). These results are comparable to values reported in the literature. The spectrofluorimetric method yielded an average filter collection efficiency of 99.9962 $\pm$ 0.0032%. The CNC method yielded the following efficiencies: average count efficiency of 99.9968 $\pm$ 0.0017% and average volume efficiency of 99.9579 $\pm$ 0.0457%. The literature reports an efficiency of >99.99%. The results are reproductible within 0.1%. (See runs 1-2, 3-4, 5-6 and 7-8). Volume collection efficiencies are obtained indirectly from the count size distribution (additional information from EC/CNC) and are therefore less accurate than efficiencies derived from count data.

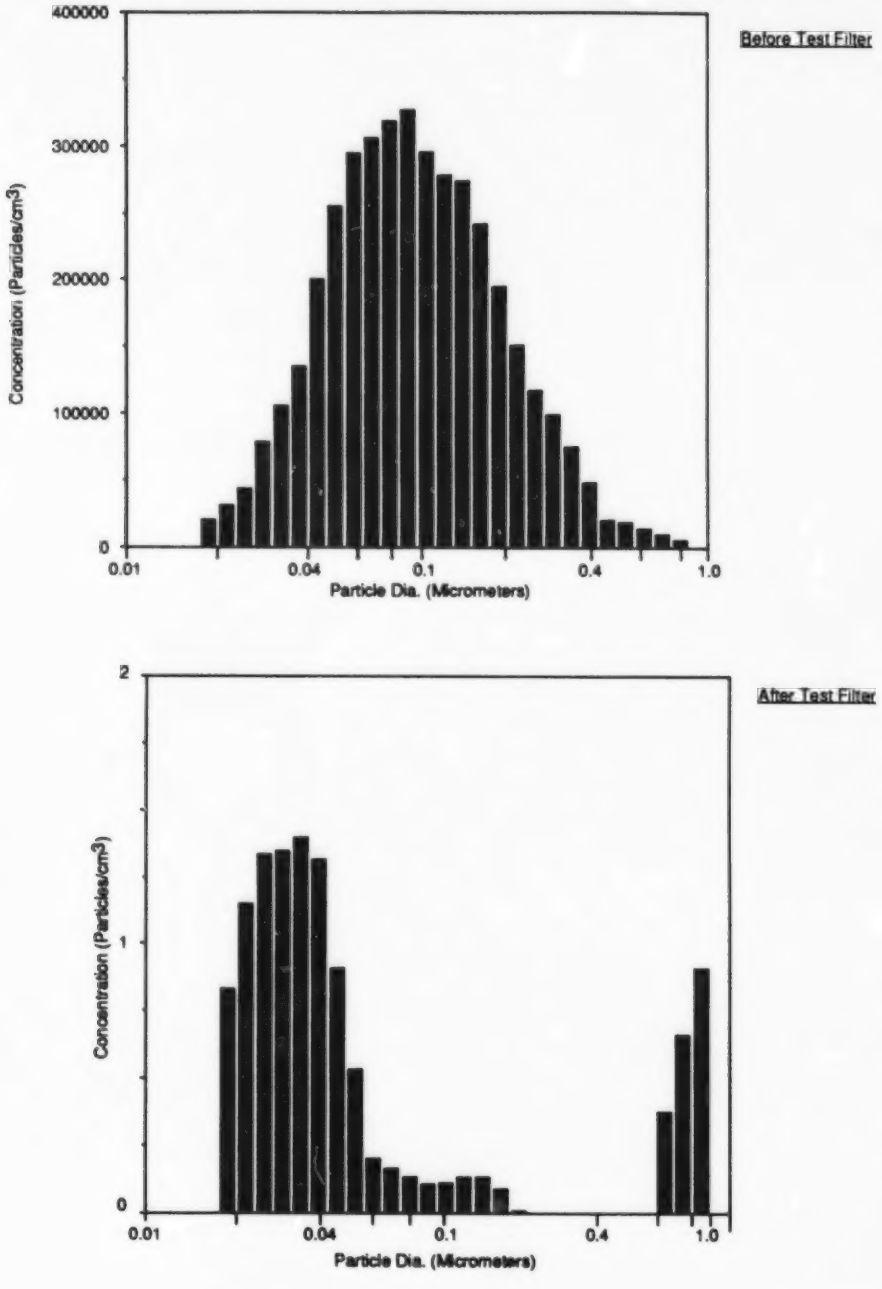

FIGURE 2

URANINE PARTICLE SIZE DISTRIBUTIONS MEASURED BEFORE AND AFTER PASSING THROUGH WHATMAN GF/B FILTER - FLOW RATE 25 L/min (RUN 1)

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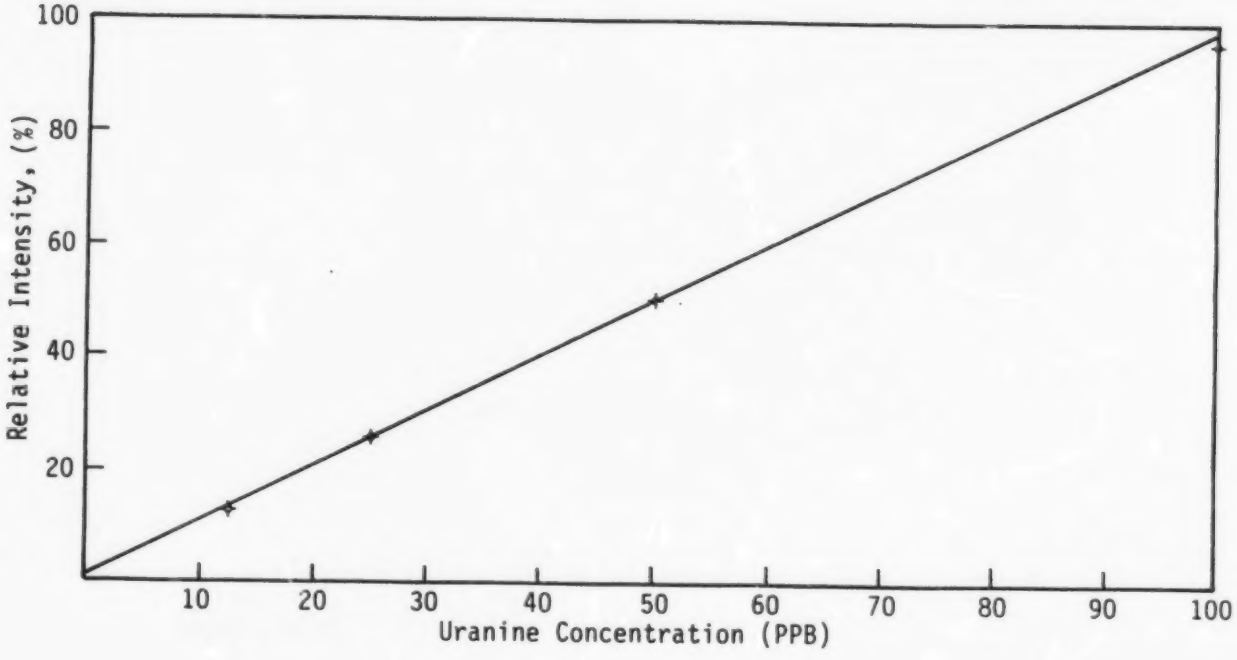

FIGURE 3

TYPICAL CALIBRATION CURVE FOR THE SPECTRO-FLUORIMETRIC ANALYSIS OF URANINE

TABLE III

RESULTS OBTAINED WITH THE SPECTROFLUORIMETRIC METHOD

Run No.*	Sampling Time (min)	Uranine Collected on Test Filter (mg)	Uranine Collected on Backup Filter (μg)	Mass Efficiency (%)
1	9	10.62	0.28	99.9974
2	9	9.18	0.50	99.9946
3	9.17	13.26	0.09	99.9993
4	9	21.12	1.20	99.9943
5	9	11.22	0.14	99.9988
6	9	12.24	0.41	99.9967
7	9	24.08	0.55	99.9977
8	9	22.44	2.10	99.9906
Mean		15.5 ± 6.0 mg (1.72 ± 0.67 mg/min)		99.9962 ± 0.0032

*See Table II

TABLE IV

RESULTS OBTAINED WITH THE CNC METHOD

Run No.*	Number Concentration (particles/cm^3) After Filter	Number Concentration (particles/cm^3) Before Filter (x 10^6)	Count Efficiency (%)	Volume Efficiency (%)	Count Mean Diameter Before Filter (μm)	Volume Mean Diameter Before Filter (μm)
1	118	3.96	99.9970	99.9744	0.0972	0.373
2	108	3.79	99.9972	99.9379	0.0954	0.362
3	139	6.94	99.9980	99.9809	0.139	0.396
4	17	9.80	99.9998	99.9978	0.133	0.358
5	164	4.21	99.9961	99.8721	0.0952	0.370
6	288	3.97	99.9927	99.9140	0.0927	0.344
7	355	11.70	99.9970	99.9886	0.119	0.319
8	475	14.30	99.9967	99.9974	0.116	0.266
		Mean	99.9968 ± 0.0017	99.9579 ± 0.0457	0.111 ± 0.018	0.349 ± 0.04

*See Table II

TABLE V

COMPARISON OF THE SPECTROFLUORIMETRIC AND CNC METHODS OF MEASURING FILTER EFFICIENCY

Run No.*	Flowrate (L/min)	CNC Method Count Efficiency (%)	CNC Method Volume Efficiency (%)	Spectrofluorimetric Method Mass Efficiency (%)	Collection Efficiency(%) Reported in the Literature+
	With Neutralizer				
1	25	99.9970	99.9744	99.9974	
2	25	99.9972	99.9379	99.9946	
3	5	99.9980	99.9809	99.9993	
4	5	99.9998	99.9978	99.9943	
	Without Neutralizer				
5	25	99.9961	99.8721	99.9988	
6	25	99.9927	99.9140	99.9966	
7	5	99.9970	99.9886	99.9977	
8	5	99.9967	99.9974	99.9906	
	Mean	99.9968 ± 0.0017	99.9579 ± 0.0457	99.9962 ± 0.0032	>99.99

* See Table II
\+ See Table I

We also examined the effects of flowrate and neutralization on filter efficiency. The results indicated that neutralization and flowrate (between 5 and 25 L/min) have no significant effect on efficiency but showed a slight effect of flowrate on particle size (Table IV). The particle count mean diameter is slightly higher in the 5 L/min run (no dilution air) than in the 25 L/min run (dilution air about 20 L/min). This may be due to incomplete drying of the aerosols by the diffusion dryer.

Since this study focused on the filter efficiency results, we did not need to determine the mass generation rate of uranine from the six-jet atomizer. However, the generation rate could be inferred from the spectrofluorimetric results (Table III). The mean uranine generation rate was about 1.72 $\pm$ 0.67 mg/min. The generation rate seems to increase with a decrease in the total air flow. Increased particle losses through the diffusion dryer at a high flowrate may partly account for the decreased generation rate. The decreased generation rate at a high flowrate may be due to a decrease in the air pressure to the atomizing nozzle, since the generation rate depends directly on the air pressure to the nozzle. The design of the six-jet atomizer has a regulated compressed air input, which subdivides into two streams - dilution air and air to the nozzle. The air pressure to the nozzle may be lowered due to increased dilution air stream. For a constant generation rate, two independent regulated compressed air sources (one for dilution and the other for air to the nozzle) are recommended.

6.0 CONCLUSION

The filter efficiency comparison study demonstrated that the CNC method for stack sampling is comparable to the presently used spectrofluorimetric method. The CNC method is preferred as it is relatively simple to operate and yields fast and continuous results.

Approved by:

O.A. Kupcis
Manager
Chemical Research Department

Prepared by:

H. Leung
Engineer
Environmental Science Section

Reviewed by:

O.T. Melo
Supervising Engineer
Environmental Science Section

M. Mackay
Student
Environmental Science Section

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